

FORM PTO-1390
(REV 12-29-99)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

Mo-6198/MA P-2.

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

To Be Assigned **09/762168**

INTERNATIONAL APPLICATION NO.

PCT/DE99/02347

INTERNATIONAL FILING DATE

July 30, 1999

PRIORITY DATE CLAIMED

August 4, 1998

TITLE OF INVENTION Continuous Method and Heat Pump Device for Enriching Liquid Low-Concentrated Reaction Mixtures

APPLICANT(S) FOR DO/EO/US BRABAND, Jurgen; MULLER, Peter; BERNHARDT, Rudiger and OTTO, Andreas

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:

Form PTO 1449 w/references
Drawing

| | | | | | |
|---|--|--|--|---|--|
| U.S. APPLICATION NO. (if known, see 37 CFR 1.5) To Be Assigned 09/762168 | | INTERNATIONAL APPLICATION NO. PCT/DE99/02347 | | ATTORNEY'S DOCKET NUMBER Mo-6198/MA P-2 | |
|---|--|--|--|---|--|

| 17. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$970.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$840.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$690.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$670.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$96.00 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div> | CALCULATIONS PTO USE ONLY | | | | | | | | | | | | | | | | | |
|---|----------------------------------|--------------|--------------|------|--------------|----------|---|-----------|--------------------|---------|---|-----------|---|--|--|------------|----|--------|
| | \$ | 860.00 | | | | | | | | | | | | | | | | |
| Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)). | \$ | 0.00 | | | | | | | | | | | | | | | | |
| <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <th style="width:15%;">CLAIMS</th> <th style="width:20%;">NUMBER FILED</th> <th style="width:20%;">NUMBER EXTRA</th> <th style="width:20%;">RATE</th> </tr> <tr> <td>Total claims</td> <td>5 - 20 =</td> <td>0</td> <td>X \$18.00</td> </tr> <tr> <td>Independent claims</td> <td>1 - 3 =</td> <td>0</td> <td>X \$78.00</td> </tr> <tr> <td colspan="3">MULTIPLE DEPENDENT CLAIM(S) (if applicable)</td> <td>+ \$260.00</td> </tr> </table> | CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | Total claims | 5 - 20 = | 0 | X \$18.00 | Independent claims | 1 - 3 = | 0 | X \$78.00 | MULTIPLE DEPENDENT CLAIM(S) (if applicable) | | | + \$260.00 | \$ | 270.00 |
| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | | | | | | | | | | | | | | | |
| Total claims | 5 - 20 = | 0 | X \$18.00 | | | | | | | | | | | | | | | |
| Independent claims | 1 - 3 = | 0 | X \$78.00 | | | | | | | | | | | | | | | |
| MULTIPLE DEPENDENT CLAIM(S) (if applicable) | | | + \$260.00 | | | | | | | | | | | | | | | |
| TOTAL OF ABOVE CALCULATIONS = | \$ | 1,130.00 | | | | | | | | | | | | | | | | |
| Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28). | \$ | 0.00 | | | | | | | | | | | | | | | | |
| SUBTOTAL = | \$ | 1,130.00 | | | | | | | | | | | | | | | | |
| Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). | \$ | 0.00 | | | | | | | | | | | | | | | | |
| TOTAL NATIONAL FEE = | \$ | 1,130.00 | | | | | | | | | | | | | | | | |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property | \$ | 0.00 | | | | | | | | | | | | | | | | |
| TOTAL FEES ENCLOSED = | \$ | 1,130.00 | | | | | | | | | | | | | | | | |
| | Amount to be refunded: | \$ | | | | | | | | | | | | | | | | |
| | charged: | \$ | | | | | | | | | | | | | | | | |

a. ☐ A check in the amount of \$_____ to cover the above fees is enclosed.


b. ☒ Please charge my Deposit Account No. 13-3848 in the amount of \$ 1,130.00 to cover the above fees.
 A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
 overpayment to Deposit Account No. 13-3848. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Noland J. Cheung
 Bayer Corporation
 Patent Department
 100 Bayer Road
 Pittsburgh, PA 15205-9741


00157
 PATENT TRADEMARK OFFICE

SIGNATURE: _____

Noland J. Cheung

NAME

39,138

REGISTRATION NUMBER

WO 00/07967

PCT/DE99/02347

1

Continuous method and heat pump device for enriching liquid low-concentrated reaction mixtures

The reaction relates to a continuous method and a heat pump device for enrichment of low-concentrated reaction mixtures resulting from the production of cycloalkanedienes by means of catalytic metathesis of cyclic aliphatic alkenes and cyclooligomers in organic reaction media.

Valuable musk odorous substances are synthetically manufactured out of cycloalkanedienes with a ring size between 12 and 18 C atoms favouring 16 C atoms, i. e. cyclohexadiene.

The production of cycloalkanedienes, especially cyclohexadecadiene is described in GB - A 1105565 (1), EP - A 0192333 (2), and EP - B 0343437 (3). After the described method, starting out from cyclic alkenes with 6 to 9 C atoms (1,2) or cyclopolyoctenylene (3) with a degree of polymerisation ≥ 3 cycloalkanedienes are formed, if parent substances of a metathesis reaction are converted in liquid phase at a supported catalyst on basis of $\text{Re}_2\text{O}_7/\text{Al}_2\text{O}_3$ (1,2) or at a modified supported catalyst.

In (2) for the first time it is mentioned, that the selectivity of target products and with that the efficiency can be increased, if the metathesis reaction does not exceed the dimerisation stage. This is possible by working with highly diluted solutions of parent substances (0.01 to 0.05 molar).

WO 00/07967

PCT/DE99/02347

2

As metathesis-inert solvents for the dilution of parents aliphatic alkanes like pentane, hexane, heptane, cyclopentane, cyclohexane, petroleum ether, chlorinated hydrocarbons like methylene chloride, chloroform, carbon tetrachloride, or aromatics like chlorobenzene and m-dichlorobenzene are used (2,3).

After flowing through the catalyst bed a low-concentrated reaction mixture in organic solvent is received containing the target product, by-products and unconverted parent substances.

According to (2,3) the reaction mixture leaving the reactor is put on a distillation device and separated into its components. This way the target product is received as relatively high-boiling fraction, while the low-boiling solvent as well as occasionally unconverted parent substances are recycled, after supplying new parent substances they are fed back into the reactor.

To increase the selectivity of the metathesis reaction in view of target products cycloalkanedienes with 12 to 18 C atoms working in highly diluted solutions with a ratio parent substance : solvent of 1:1300 is absolutely required.

This fact is the essential disadvantage of this method, till now considerably limiting or preventing the technical realisation of the method. Separating the reaction solution by means of batch distillation as usual into target product and solvent requires high energy for evaporation of the solvent and increases the costs for the production of the target product and this way also the price of the synthetic odorous substance considerably.

WO 00/07967

PCT/DE99/02347

3

Thus, the task is to find a method to lower the energy costs for the separation of the reaction solution into target product, by-products, and organic reaction medium (solvent) considerably and to recycle the organic reaction medium at the same time.

Now a method has been found to solve the above mentioned task by applying the principle of a heat pump in such a way, that the heat level of the organic reaction medium is permanently increased and decreased within a narrow temperature range and no permanent evaporation and condensation is required. This target is achieved by means of a suitable connection of heat exchangers in combination with a vent compressor. Electric energy is converted into heat via a vent compressor, thus the organic reaction medium is compressed and heated. The heat absorbed in the vent compressor is directly applied for evaporating the organic reaction medium out of the prevailing reaction mixture in the heat exchanger, thus a new cycle starts.

While elements of the method are known the combination of these elements as an inventive method so far has not been applied in odorous substance chemistry.

The method to resolve above mentioned task is characterised by the features of claims 1 to 4 and can be executed by means of the device described in claim 5.

The low-concentrated reaction mixture in the organic reaction medium is continuously fed via a feeding line (1) into a single or multiple chambered condenser with temperature T1.

WO 00/07967

PCT/DE99/02347

4

This temperature is determined by the reaction conditions of the catalytic metathesis reaction, especially by the infusibility of the catalyst, the selectivity of the catalytic reaction with regard to conversion of parent substances to the target product depending on temperature and the organic reaction medium applied in connection with that. Boiling points of aliphatic, cyclic aliphatic, and chlorinated hydrocarbons stated in claim 2 are in a range between 30°C and 120°C. With regard to the aim to keep energy expenses for the enrichment of the reaction mixture in the organic reaction medium as low as possible the organic reaction medium has to be chosen in such a way, that its boiling point is just a few centigrade above the reaction temperature of parent substances at the catalyst.

For that reason the organic reaction medium has to be chosen in such a way the temperature difference between entry temperature T1 of the reaction solution in the evaporator and the boiling point of the organic reaction medium to be evaporated does not exceed 5 K.

By enriching the reaction mixture in the organic reaction medium the boiling point of the applied solvent is increasing. The boiling point curve shows, that the boiling point of the solvent increases significantly, when the reaction mixture in the organic reaction medium is enriched to more than 50 w/w %. Depending on actual conditions of the method the energetic demand is lowest when the reaction mixture in the evaporator is enriched in the organic reaction medium to 30 to 50 w/w %.

From the evaporator the vapours of the organic reaction medium are sucked off and compressed, which raises the temperature to T2. The pressure difference is chosen in such a way, that the temperature T2 is 5 to 12 K above T1.

This is achieved by a pressure difference of 0.25 to 1.0 bar. The vapour of the organic reaction medium compressed to temperature T2 is fed back to the heat exchanger of the evaporator and transfers the absorbed heat energy to the organic reaction medium being in the evaporator at temperature T1, which, as a result, is evaporating. This heat exchange is most economical, if the temperature difference between T1 and T2 does not fall below 5 K, preferably between 8 and 12 K.

Applying the described method low-concentrated liquid reaction mixtures with a content of 0.1 w/w % at least in the organic reaction medium are enriched with lower energy demand to 30 to 50 w/w % in the evaporator.

The reaction mixture enriched to that percentage of valuable product is permanently sucked off via pipe (9) from the evaporator and fed to the distillation plant (10) for further separation into organic reaction medium, unconverted parent substances, by-products and target product. Organic reaction medium and unconverted parent substances are again unified in vessel (8) with the organic reaction medium condensed in the heat exchanger (6) of the evaporator (2), added to parent substances and fed into the reactor.

Applying the inventive method energy consumption for the evaporation of the organic reaction medium can be lowered by 80 %, expenses by about 30%. The following example will explain the method practically.

WO 00/07967

PCT/DE99/02347

6

Example

A reaction mixture from the reactor with a content of valuable product of 0.34 w/w % containing 2 w/w % of cyclooctene, 30 w/w % of cyclohexadecadiene, and 60 w/w % of oligomers in n-hexane is continuously fed into a multiple-chamber evaporator. At an evaporator efficiency of about 17,000 kg/h n-hexane the content of valuable product mixture in the evaporator is increased to 35 w/w %.

The entry temperature amounts to 67°C. The boiling point of n-hexane amounts to 68.5°C. By increasing the concentration of the valuable product mixture in n-hexane up to 35 w/w % the boiling point of the mixture in the evaporator increases to 72°C (T1). In the compressor n-hexane vapour is compressed by means of a pressure difference of 0.41 bar which increases the temperature to 81°C (T2). This way T2 minus T1 amounts to 9 K.

The compressed n-hexane vapour is lead through the heat exchangers of the evaporator and transfers heat energy to n-hexane of temperature T1. After that condensed n-hexane goes from heat exchanger into the mixture vessel.

At the above mentioned evaporation of n-hexane continuously 165 kg/h of reaction mixture enriched to 35 w/w % are sucked off from the evaporator. The share of the valuable product mixture is 57.5 kg. Energy consumption is about 100 kW/h.

Claims for a patent

- 1 Continuous method for enrichment of reaction mixtures with a concentration of 0.1 w/w % at least consisting of unconverted parent substances, the target product, and by-products resulting from the catalytic metathesis of cycloalkanedienes from mixtures of cyclic aliphatic alkenes and cyclooligomers in liquid organic reaction media being aliphatic, cyclic aliphatic and chlorinated hydrocarbons, in which the liquid organic reaction media and the unconverted part of parent substances are recycled, for the use as heat transfer medium at a temperature difference of 5 K at least (T_1/T_2) evaporated and compressed, and finally are fed in condensed state to a mixing vessel for adjusting the concentration ratios of parent substances with regard to the method, covering all measures of the method.
 - 1a Feeding of low-concentrated reaction mixtures with temperature T_1 withdrawn continuously from the reactor into a single or multiple-chambered evaporator.
 - 1b Sucking off of vapours of the organic reaction medium from the evaporator.
 - 1c Compressing of vapours in a vent compressor by means of feeding of electric energy.
 - 1d Feedback of compressed vapours with temperature T_2 into the heat exchanger of the evaporator.

WO 00/07967

PCT/DE99/02347

8

- 1e Heat exchange in the evaporator between the low-concentrated reaction mixture of temperature T1 and the compressed organic reaction medium of temperature T2 fed back.
- 1f Continuous feedback of condensed organic reaction medium from the heat exchanger of the evaporator into a mixing vessel for adjusting the concentration ratios with regard to the method.
- 1g Continuous withdrawing of the reaction mixture enriched in the reaction medium to a content of 30 w/w % at least from the evaporator consisting of target product, by-products, parent substances, and organic reaction medium.
- 1h Feeding the enriched reaction mixture into a high-grade distillation plant to distil off the residual part of the organic reaction medium and to separate the reaction mixture into target product, by-products, and parent substances.
- 1i Feedback of condensed organic reaction medium and parent substances to the mixing vessel according to 1f.
- 2 Method according to claim 1 characterised by the fact, that under aliphatic, cyclic aliphatic, and chlorinated hydrocarbons pentane, hexane, heptane, cyclopentane, cyclohexane, cycloheptane, methylene chloride, chloroform, carbon tetrachloride, and petroleum ether are understood.
- 3 Method according to claim 1 and 2 characterised by the fact, that temperature T1 does not fall more than 5 K below of the boiling point of the liquid organic reaction medium.

WO 00/07967

PCT/DE99/02347

9

- 4 Method according to claim 1 to 3 characterised by the fact, that the temperature difference between T1 and T2 amounts preferably to 8 to 12 K.
- 5 Device to carry out the method according to claim 1 to 4 consisting of
- 5a a feeding pipe (1) for low-concentrated reaction mixture in liquid organic reaction medium,
- 5b a single or multiple-chambered evaporator (2) including heat exchanger (6), what the liquid organic reaction medium evaporates in and the other components of the reaction mixture (target product, by-products, parent substances) are enriched,
- 5c a suction pipe (3) to feed the vapour of the organic reaction medium to the compressor (4),
- 5d a compressor (4) to compress the vapour of the organic reaction medium and to heat it to temperature T2,
- 5e a pressure pipe (5) to feed the compressed and heated organic reaction medium to the heat exchanger (6),
- 5f a heat exchanger (6) inside the evaporator (2) for transfer of heat between the liquid organic reaction medium as well as the reaction mixture with temperature T1 and the compressed organic reaction medium with temperature T2,

WO 00/07967

PCT/DE99/02347

10

- 5g a pipe (7) to carry the condensed organic reaction medium from heat exchanger (6) to mixing vessel (8),
- 5h a pipe (9) to carry the enriched reaction mixture from evaporator (2) to high-grade distillation plant (10),
- 5i a high-grade distillation plant (10) for the separation of the enriched reaction mixture into target product, by-products, parent substances, and organic reaction medium, and
- 5j a pipe (11) to carry the liquid organic reaction medium from distillation plant (10) to mixing vessel (8).

Flow sheet enclosed

Abstract

The invention relates to a continuous method and a heat pump device for enrichment of low-concentrated reaction mixtures resulting from the production of cycloalkanedienes by means of catalytic metathesis of cyclic aliphatic alkenes and cyclooligomers in organic reaction media.

Using the heat pump principle, liquid reaction mixtures with a content of ≥ 0.1 w/w % are enriched in an organic reaction medium to 30 to 50 w/w %. In the course of that, the organic reaction medium of temperature T_1 is evaporated in the evaporator, the vapour is sucked off and compressed to temperature T_2 in a compressor, whose pressure difference ranges from 0.25 to 1 bar.

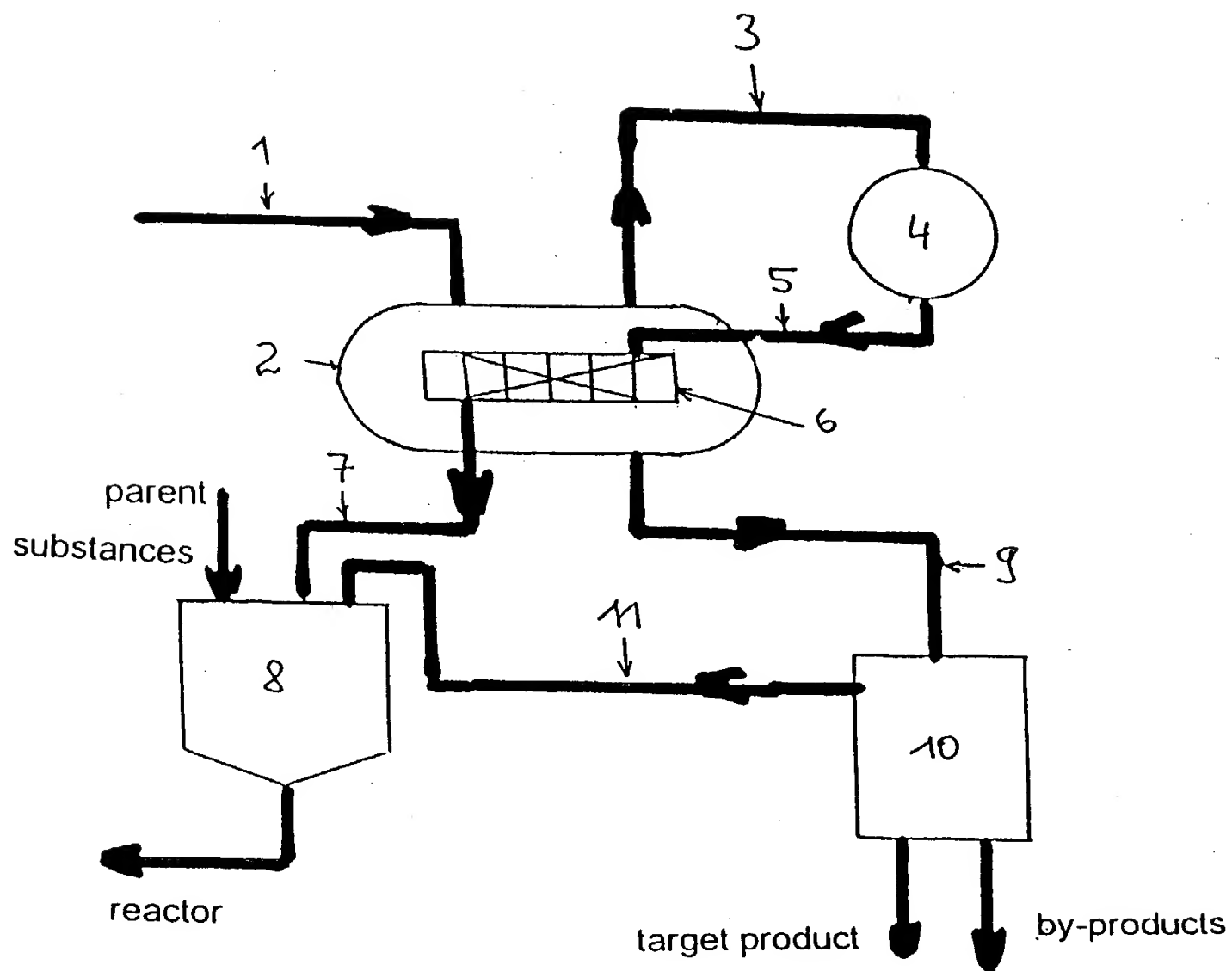
The compressed vapour of the reaction medium transfers the heat energy obtained from electric energy in the heat exchanger of the evaporator to the organic reaction medium of temperature T_1 , but the temperature difference T_2 minus T_1 must not exceed 12 K.

After the inventive method, energy consumption for evaporation of the organic reaction medium is reduced by about 80 % and costs by about 30 %.

FIGURE 1**Legend:**

- 1 feeding pipe
- 2 evaporator
- 3 suction pipe
- 4 compressor
- 5 pressure pipe

- 6 heat exchanger
- 7 pipe
- 8 mixing vessel
- 9 pipe
- 10 high-grade distillation plant
- 11 pipe





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#3

Attorney Docket No. _____

COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office and address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Continuous method and heat pump device for enrichment of low-
concentrated reaction mixtures resulting from the production of
cycloalkanediene in liquid organic reaction media

the specification of which (check one)

- ☐ is attached hereto ☐ was filed on _____ as Application
Serial No. _____ and was amended on
_____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with title 37, Code of Federal Regulations, §1.56 (a).

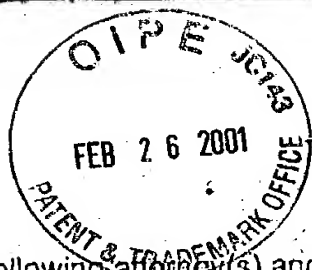
I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below and foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

PRIOR FOREIGN APPLICATION(S)

| | | | |
|-------------------|-----------|------------------------|---|
| <u>198 35 203</u> | <u>DE</u> | <u>04. Aug. 1998</u> | Priority claimed |
| (Number) | (Country) | (Day/month/year filed) | <input checked="" type="checkbox"/> Yes <input type="checkbox"/> No |
| _____ | _____ | _____ | <input type="checkbox"/> Yes <input type="checkbox"/> No |
| (Number) | (Country) | (Day/month/year filed) | |
| _____ | _____ | _____ | <input type="checkbox"/> Yes <input type="checkbox"/> No |
| (Number) | (Country) | (Day/month/year filed) | |

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT International filing date of this application:

| | | |
|--------------------------|----------------------|--------------------------------------|
| <u>PCT/DE 99/02347</u> | <u>30. July 1999</u> | <u>patented</u> |
| (Application Serial No.) | (Filing Date) | (STATUS: patented/pending/abandoned) |
| _____ | _____ | _____ |
| (Application Serial No.) | (Filing Date) | (STATUS: patented/pending/abandoned) |



09762163 .022601

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office Connected therewith:

JOSEPH C. GIL, Patent Office Registration Number 26,602
ARON PREIS, Patent Office Registration Number 20,426
LYNDANNE M. WHALEN, Patent Office Registration Number 29,457
THOMAS W. ROY, Patent Office Registration Number 29,582
RICHARD E.L. HENDERSON, Patent Office Registration Number 31,619
GODFRIED R. AKORLI, Patent Office Registration Number 28,779
N. DENISE BROWN, Patent Office Registration Number 36,097
NOLAND J. CHEUNG, Patent Office Registration Number 39,138
DIDERICO VAN EYL, Patent Office Registration Number 38,641
CAROLYN M. SLOANE, Patent Office Registration Number 44,339
JAMES R. FRANKS, Patent Office Registration Number 42,552
JACKIE ANN ZURCHER, Patent Office Registration Number 42,251

all of Bayer Corporation, Pittsburgh, Pennsylvania 15205-9741

SEND CORRESPONDENCE TO:

Patent Department
Bayer Corporation
100 Bayer Road
Pittsburgh, Pennsylvania 15205-9741

DIRECT TELEPHONE CALLS TO:
(Name and telephone number)

Patent Department
(412) 777-2349

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1-00 1. FULL NAME OF FIRST INVENTOR: Dr. Braband, Jürgen

RESIDENCE: D-04275 Leipzig

POST OFFICE ADDRESS: Hardenbergstr. 44

CITIZENSHIP: DE, Germany

DATUM: 1/23/01

INVENTOR'S SIGNATURE:

Jürgen Braband

2-00 2. FULL NAME OF SECOND JOINT INVENTOR; IF ANY: Dr. Müller, Peter

RESIDENCE: D-04175 Leipzig

POST OFFICE ADDRESS: Nonnenstr. 5a

CITIZENSHIP: DE, Germany

DATUM: 1/23/01

INVENTOR'S SIGNATURE:

Peter Müller

3-00 3. FULL NAME OF THIRD JOINT INVENTOR, IF ANY: Bernhardt, Rüdiger

RESIDENCE: D-06774 Plodda

POST OFFICE ADDRESS: Schmerzner Weg 4

CITIZENSHIP: DE, Germany

DATUM: 1/23/01

INVENTOR'S SIGNATURE:

Rüdiger Bernhardt

Page 2

4-00

4. FULL NAME OF FOURTH JOINT INVENTOR, IF ANY: Otto, Andreas

RESIDENCE: D-06800 Jessnitz

POST OFFICE ADDRESS: Nordstr. 31

DEX

CITIZENSHIP: DE, Germany

DATUM: 1/23/01

INVENTOR'S SIGNATURE:

Andreas Otto